

## \* NOTICES \*

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## CLAIMS

(57) (Claim 6)

(Claim 1)

A polyoxyalkylene series polymer (A) which has a silicon containing functional group which can construct a bridge by forming a siloxane bond,

Silicate (B),

an amine compound — and

As a silanol condensation catalyst, they are carboxylic acid tin salt (C) and a non-tin catalyst (E).

It is a hardenability constituent to contain,

Silicate (B) is tetra alkoxysilane or its partial hydrolysis condensate,

A carbon atom in which carboxylic acid tin salt (C) adjoins a carbonyl group is the carboxylic acid tin salt which is the 4th class carbon,

A carbon atom in which a non-tin catalyst (E) adjoins a carbonyl group is carboxylic acid which is the 4th class carbon.

A hardenability constituent.

(Claim 2)

A polyoxyalkylene series polymer in which a polyoxyalkylene series polymer (A) which has a silicon containing functional group which can construct a bridge by forming a siloxane bond introduced an unsaturation group into an end, and a general formula (1):



an allyl group of the carbon numbers 1-20 from which R<sup>1</sup> in a formula and R<sup>2</sup> were the same as or different. When the TETRAORGANO siloxy group shown by any group of the carbon numbers 6-20, an allyl group of the carbon numbers 7-20, or (R<sup>1</sup>)<sub>3</sub>SiO— is shown and R<sup>1</sup> or two or more R<sup>2</sup> exist, they may be the same and may differ. R<sup>1</sup> is a hydrocarbon group of monovalence of the carbon numbers 1-20 here, and three R<sup>2</sup> may be the same and may differ. X shows a hydroxyl group or a hydroxyloxy group, and when two or more X exists, they may be the same and may differ. As for a, 0, 1, 2, or 3b shows 0, 1, or 2, respectively. About b in m bases (SiR<sup>1</sup>)<sub>2</sub>-X<sup>1</sup>-X<sup>2</sup>-O, they may be the same and may differ. m shows an integer of 0 to 10, however — what satisfies a-sigma b>1 — carrying out — the hardenability constituent according to claim 1, being a polyoxyalkylene series polymer obtained by an addition reaction with a hydrosilane compound expressed.

(Claim 3)

The hardenability constituent according to claim 1 or 2 in which a polyoxyalkylene series polymer (A) which has a silicon containing functional group which can construct a bridge by forming a siloxane bond is characterized by being a polymer which does not contain an amide segment (-NH-CO-)

understandably in a principal chain skeleton.

(Claim 4)

An additive for interior panels containing a hardenability constituent of a description in any 1 paragraph of Claims 1-3, as an object for face panels, or adhesives for car panels.

(Claim 5)

A sealing material for working joint of a building containing a hardenability constituent of a description in any 1 paragraph of Claims 1-3.

[Translation done.]

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## DETAILED DESCRIPTION

[Detailed Description of the Invention]

[0001]

[Field of the Invention]

This invention relates to the hardenability constituent containing the organic polymer which has a silicon containing functional group (hereinafter, a reactive silicon group) which can construct a bridge by forming a siloxane bond.

[0002]

[Description of the Prior Art]

It is known that the organic polymer which contains at least one reactive silicon group in a molecule thereof is used as a material which constructs a bridge by formation of the siloxane bond accompanied by the hydrolysis reaction of a reactive silicon group, etc., and a rubber-like hardened material is obtained with hygroscopic surface moisture, etc., also in a room temperature.

[0003]

In the polymer which has these reactive silicon groups, a polyoxymethylene series polymer and a polyoxymethylene system polymer are already produced industrially, and are widely used for uses, such as a sealing material, adhesives, and a paint.

[0004]

The adhesives for interior panels, the adhesives for face panels, the adhesives for tiling, the adhesives for stone tensions, when the resin for adhesives used for the adhesives for finishing of wall, the adhesives for car panels, etc., is inferior to stability or creep resistance, an adhesives layer may pass with prudence and the stress from the outside of adherent, it may change by the time, and a panel tile, a stone, etc., may shift. Also in ceiling finishing adhesives or floor finishing adhesives, if inferior to stability or creep resistance, an adhesives layer may pass and it may change by the time, and unevenness of a ceiling surface or a floor line may arise. If the stability of the electrical and electric equipment, an electron, and the adhesives for precision-mechanical-equipment assemblies and creep resistance are bad, an adhesives layer may pass, and it may change by the time, and may be connected with the degradation of apparatus. Therefore, it is called for that the constituent for these adhesives is excellent in stability or creep resistance.

[0005]

The sealing material generally fills up the joined part and crevice between various members, and he is used in order to give watertight and airtightness. Therefore, since the material returns to the use part over a long period of time is very important, excellent in stability or endurance is called for as physical properties of a hardened material. Working joint of a building with an especially large change of joint width (Kasag) the circumference of glass, the circumference of a window frame and a sash, a curtain wall, and various face panels — business — stability and endurance excellent in the constituent used for a sealing material, the sealing material for direct grouting, the sealing material for multiple glass, the sealing material for speed signal generator construction methods, etc., are called for.

[0006]

On the other hand, (the patent documents 1), the (patent documents 2), the (patent documents 3), the (patent documents 4), (The patent documents 5), the (patent documents 6), the (patent documents 7), the (patent documents 8), (The patent documents 9), the (patent documents 10), the (patent documents 11), the (patent documents 12), (The patent documents 13), the (patent documents 14), the (patent documents 15), the (patent documents 16), (The patent documents 17),

[Patent documents 1]  
JP-H10-245482A

[Patent documents 2]  
JP-H10-246484A

[Patent documents 3]  
JP-H10-251552A

[Patent documents 4]  
JP-H10-324793A

[Patent documents 5]  
JP-H10-330630A

[Patent documents 6]  
JP-H10-124753A

[Patent documents 7]  
JP-H11-124804A

[Patent documents 8]  
JP-H11-21463A

[Patent documents 9]  
JP-H11-29713A

[Patent documents 10]  
JP-H11-49869A

[Patent documents 11]  
JP-H11-49970A

[Patent documents 12]  
JP-H11-118631A

[Patent documents 13]  
JP-H11-124509A

[Patent documents 14]  
WO No. 47839 [ 88 to ]

[Patent documents 15]  
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[Patent documents 16]  
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they may be the same and may differ, R' is a hydrocarbon group of monovalence of the carbon numbers 1-20 here, and three R may be the same and may differ. X shows a hydroxyl group or a hydroxylic basis, and when two or more X exists, they may be the same and may differ. As for a, 0, 1, 2, or 3b shows 0, 1, or 2, respectively. About b in m based (SiR<sub>2</sub>-2-X<sub>2</sub>-O) may be the same and may differ, m shows an integer of 0 to 19, however — what satisfies a+3m b=1 — carrying out — it is related with stability, endurance, and creep resistance corrective strategy of a hardened material given in said either using a hardenability constituent which is an organic polymer obtained by an addition reaction with a hydrosilane compound expressed.

An organic polymer in which an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment introduced an unsaturation group into an end, and general formula (2) :



X in a formula shows a hydroxyl group or a hydroxylic basis, and three X may be the same and it may differ. It is related with stability, endurance, and creep resistance corrective strategy of a hardened material given in said either using a hardenability constituent which is an organic polymer obtained by an addition reaction with a hydrosilane compound expressed.

[0048] The 4th is an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond in said invention, and this organic polymer is a general formula (3) :

$$\text{O-R}^2\text{-CH(OR}^1\text{)-CH}_2\text{-(SiR}^2\text{)-2-X}_2\text{-O} \quad \text{m-SiR}^2\text{-2-X}_2\text{-X}_3 \quad (3)$$

(R<sup>1</sup> formula) a divalent organic group of the carbon numbers 1-20 which contain one or more sorts chosen from a group which consists of hydrogen, oxygen, and nitrogen as a composition atom [ show and ] R<sup>1</sup>, R<sup>2</sup>, X, a, b, and m — the above — it is the same — it is related with stability, endurance, and creep resistance corrective strategy of a hardened material using a hardenability constituent, which is an organic polymer (A3) which has a structure part with which it is expressed.

[0049] An organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment is a general formula (4) :

$$\text{O-R}^2\text{-C(OR}^1\text{)-CH}_2 \quad (4)$$

An organic polymer which introduced an unsaturation group expressed with (R<sup>2</sup> is the same as the above), and general formula (1) :

$$\text{H-SiR}^2\text{-2-X}_2\text{-O} \quad \text{m-SiR}^2\text{-2-X}_2\text{-X}_3 \quad (1)$$

It is related with stability, endurance, and creep resistance corrective strategy of a hardened material given in the above using a hardenability constituent which is an organic polymer obtained by an addition reaction with a hydrosilane compound expressed with (R<sup>1</sup> in formula, R<sup>2</sup>, X, a, b, and m are the same as the above).

[0050] An organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment is a general formula (5) :

$$\text{O-R}^2\text{-CH(OR}^1\text{)-CH}_2\text{-SiX}_3 \quad (5)$$

(R<sup>1</sup> formula and X are the same as the above) — it is related with stability, endurance, and creep resistance corrective strategy of a hardened material given in said either using a hardenability constituent which is an organic polymer which has a structure part with which it is expressed.

[0051] An organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment, it is related with stability, endurance, and creep resistance corrective strategy of a hardened material given in said either using for a principal chain skeleton a hardenability constituent which is an organic polymer which does not contain an amide segment (—NH—CO—) substantially.

[0052]

A silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment is a general formula (6) :

$$\text{-(SiOR}^1\text{)}_3 \quad (6)$$

(three R<sup>1</sup> is an organic group of monovalence of the carbon numbers 2-20 independently among a formula, respectively) — it is related with stability, endurance, and creep resistance corrective strategy of a hardened material given in said either using a hardenability constituent which is a basis expressed.

[0053] It is related with stability, endurance, and creep resistance corrective strategy of a hardened material given in said either, wherein a silicon containing functional group which can construct a bridge by forming a siloxane bond uses a hardenability constituent which is a triethoxy silyl group as a desirable embodiment.

[0054]

The 5th is an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of this invention. A silicon containing functional group which can construct a bridge by forming a siloxane bond is related with this layer hardenability corrective strategy using an silicon an organic polymer (A1) which is a silicon containing functional group which consists of three or more hydroxylic bases, and a hardenability constituent containing an organic tin catalyst (D).

[0055]

The 6th is an organic polymer (A1) which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of this invention. Adhesives for interior panels containing silicate (B). Adhesives, it is related with floor finishing adhesives, adhesives for stone tensions, ceiling finishing adhesives, it is related with floor finishing adhesives, adhesives for finishing of wall.

adhesives for car panels, the electrical and electric equipment, an electron and adhesives for precision-mechanical-equipment assemblies, a sealing material for direct graining a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0056]

The 7th is an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of this invention. A silicon containing functional group which can construct a bridge by forming a siloxane bond, it is characterized by containing an organic polymer (A1) which is a silicon containing functional group which has three or more hydroxylic bases on silicon. Adhesives for interior panels, adhesives for floor panels, adhesives for tiling adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, it is related with the electrical and electric equipment, an electron and adhesives for precision-mechanical-equipment assemblies, a sealing material for direct graining a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0057]

A main chain of an organic polymer (A1) which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment. Using a hardenability constituent which is an acrylic ester system copolymer manufactured by a living radical-polymerization method (meta) to the above by which it is characterized A description. Adhesives for interior panels, adhesives for floor panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, it is related with the electrical and electric equipment, an electron and adhesives for precision-mechanical-equipment assemblies, a sealing material for direct graining, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0058]

As a desirable embodiment, containing silicate (B) further to said either by which it is characterized A description. Adhesives for interior panels, adhesives for floor panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, it is related with the electrical and electric equipment, an electron and

adhesives for precision-mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0059]

As a desirable embodiment, containing carboxylic acid an salt (O) further to said either by which it is characterized A description, Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0060]

As a desirable embodiment, containing an organic tin catalyst (D) further to said either by which it is characterized A description, Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0061]

The 8th is an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of the invention. It is characterized by this organic polymer being an organic polymer (A2) which averages per molecule a silicon containing functional group which can construct a bridge by forming a siloxane bond and has it 1.7-5 pieces. Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0062]

A silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment, That it is a silicon containing functional group which has three or more hydroxylic bases on the above by which it is characterized A description, Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0063]

An organic polymer in which an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment introduced an unsaturation group into an end and a general formula (1):



That it is an organic polymer obtained by an addition reaction with a hydrosilane compound expressed with (R<sup>1</sup> in terms of R<sup>2</sup>, X, a, b, and m are the same as the above) to said either by which it is characterized A description, Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0064]

An organic polymer in which an organic polymer which has a silicon containing functional group which

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can construct a bridge by forming a siloxane bond as a desirable embodiment introduced an unsaturation group into an end and a general formula (2):



That it is an organic polymer obtained by an addition reaction with a hydrosilane compound expressed with (X in a formula is the same as the above) to said either by which it is characterized A description, Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0065]

The 9th is an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of the invention, and this organic polymer is a general formula (3):



It is characterized by being an organic polymer (A3) which has a structure part expressed with (R<sup>1</sup> in terms of R<sup>2</sup>, X, a, b, and m are the same as the above), Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0066]

An organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment is a general formula (4):



An organic polymer which introduced an unsaturation group expressed with (R<sup>1</sup> is the same as the above), and general formula (1):



That it is an organic polymer obtained by an addition reaction with a hydrosilane compound expressed with (R<sup>1</sup> in terms of R<sup>2</sup>, X, a, b, and m are the same as the above) to the above by which it is characterized A description, Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0067]

An organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment is a general formula (5):



(R<sup>1</sup> in terms of X are the same as the above) — a description to said either being an organic polymer which has a structure part with which it is expressed, Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tensions, Ceiling finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels, It is related with the electrical and electric equipment, an electron and mechanical-equipment assemblies, a sealing material for direct grating, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0068]

An organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment, That it is an organic polymer which does not

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contain an amide segment  $(-NH-CO-)$  substantially in a principal chain skeleton to said other by which it is characterized. Adhesives for interior panels, adhesives for face panels, adhesives for tiling, adhesives for stone tiles, caulking finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels. It is related with the electrical and electric equipment, an electron and adhesives for precision-mechanical-equipment assemblies, a sealing material for direct grouting, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

A silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment is a general formula (6):



( $R^{1n}$  formula is the same as the above.) — a description to said other being a basis expressed, Adhesives for interior panels, adhesives for face panels, adhesives for tiling adhesives for stone tiles, caulking finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels. It is related with the electrical and electric equipment, an electron and adhesives for precision-mechanical-equipment assemblies, a sealing material for direct grouting, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

A silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment is a general formula (7):  
It is a triethoxy group, in which it is characterized by having three ethoxy groups, adhesives for interior panels, adhesives for face panels, adhesives for tiling adhesives for stone tiles, caulking finishing adhesives, floor finishing adhesives, adhesives for finishing of wall, adhesives for car panels. It is related with the electrical and electric equipment, an electron and adhesives for precision-mechanical-equipment assemblies, a sealing material for direct grouting, a sealing material for multiple glass, a sealing material for speed signal generator construction methods, or a sealing material for working joint of a building.

[0071]

The 10th is an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of this invention. A silicon containing functional group which can construct a bridge by forming a siloxane bond is related with an organic polymer (A1) which is a silicon containing functional group which has three or more hydroxylic bases on silicon, and a hardenability constituent containing silicate (5).

[0072]

As a desirable embodiment, silicate is related with a hardenability constituent given in the above being a condensate of tetra siloxylane.

[0073]

The 11th is an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of this invention. A silicon containing functional group which can construct a bridge by forming a siloxane bond is related with an organic polymer (A1) which is a silicon containing functional group which has three or more hydroxylic bases on silicon, and a hardenability constituent containing a carboxylic acid salt (G1) whose carbon of an alpha position of the carboxylic group is the 4th class carbon.

[0074]

The 12th is an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of this invention. A silicon containing functional group which can construct a bridge by forming a siloxane bond is related with an organic polymer (A1) and carboxylic acid salt (G) which are the silicon containing functional groups which have three or more hydroxylic bases on silicon, and a hardenability constituent containing an organic tin catalyst (D).

[0075]

The 13th is an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of this invention. A silicon containing functional group which can construct a bridge by forming a siloxane bond is related with an organic polymer (A1) which is a silicon containing functional group which has three or more hydroxylic bases on silicon, and a hardenability constituent containing a non-tin catalyst (E).

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[0076]

The 14th is an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of this invention. A silicon containing functional group which can construct a bridge by forming a siloxane bond is related with an organic polymer (A1) which is a silicon containing functional group which has three or more hydroxylic bases on silicon, and a hardenability constituent containing a minute hollow body (F).

[0077]

The 15th is an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of this invention. A silicon containing functional group which can construct a bridge by forming a siloxane bond. It is a hardenability constituent containing an organic polymer (A1) which is a silicon containing functional group which has three or more hydroxylic bases on silicon, and is related with a hardenability constituent, wherein this organic polymer is 5 to 28 % of the weight in a total amount of a hardenability constituent.

An organic polymer in which an organic polymer which has a silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment introduced an unsaturation group into an end, and general formula (2):  
 $H-Si(R^{2n})_3 \quad (2)$

It is related with a hardenability constituent given in said other being an organic polymer obtained by an addition reaction with a hydrosilane compound expressed with  $OX$  in a formula is the same as the above.

[0078]

A silicon containing functional group which can construct a bridge by forming a siloxane bond as a desirable embodiment is a general formula (6):



It is related with a hardenability constituent given in said other being a basis expressed with ( $R^{2n}$  formula is the same as the above).

[0080]

A silicon containing functional group which has a silicon containing functional group which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of this invention, and can construct a bridge by forming a siloxane bond is a general formula (6):



An organic polymer (A4) which has a basis expressed with ( $R^{2n}$  formula is the same as the above), and general formula (7):  
 $-SiR^5_3 \quad (OR^5)_3 \quad (7)$

( $R^5$  is an organic group of monovalence of the carbon numbers 1-20 independently among a formula, respectively, and 3- $\infty$   $R^5$ ) It is an organic group of monovalence of the carbon numbers 2-20 independently, and shows 0, 1, or 2, respectively. It is related with a hardenability constituent in which is improved, a hardenability constituent containing an amine silane coupling agent (G) which has a basis expressed has been improved.

[0081]

A silicon containing functional group which has a silicon containing functional group which has a silicon containing functional group which can construct a bridge by forming a siloxane bond of this invention, and can construct a bridge by forming a siloxane bond is a general formula (6):



An organic polymer (A4) which has a basis expressed with ( $R^{2n}$  formula is the same as the above), and general formula (8):  
 $-SiR^6_3 \quad (OR^6)_3 \quad (8)$

( $R^6$  is an organic group of monovalence of the carbon numbers 1-20 independently among a formula, respectively,  $R^6$  of a 3- $\infty$  individual is an organic group of monovalence of the carbon numbers 2-20 independently, respectively, shows 0, 1, or 2 and a basis expressed 1, 2, or 3.) However, 3- $\infty$

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dioxyacetate and xylylene diacrylate; isophore diisocyanate. What is obtained from a reaction of polyisocyanate compounds, such as various isocyanate system polyisocyanates, such as hexamethylene diisocyanate, and polyol which has various kinds of above-mentioned principal chain skeletons can be a hardener.

[0096]

If there are many amide segments (—NH—CO—) generated in a principal chain skeleton based on said urethane bond, viscosity of an organic polymer will become high and will serve as a bad constituent of workability. Therefore, as for quantity of an amide segment occupied in a principal chain skeleton of an organic polymer, it is preferred that it is 3 or less of the weight, it is more preferred that it is 1 or less of the weight, and it is most preferred that an amide segment is not included substantially.

[0097]

A reactive silicon group contained in an organic polymer which has a reactive silicon group is a basis which can construct a bridge by forming a siloxane bond by the reaction which has a hydroxyl group or a hydrolytic basis combined with a silicon atom, and is accelerated by a silanol condensation catalyst. As a reactive silicon group, it is a general formula (11):



an alkyl group of the carbon numbers 1-20 from which  $R^1$  in formula and  $R^2$  were the same or different. When the Tert ORGANO siloxy group shown by aryl group of the carbon numbers 6-20, an aralkyl group of the carbon numbers 7-20, or (R) $_3$ SiO $_2$  is shown and  $R^1$  or two or more  $R^2$  exist, they may be the same and may differ.  $R^1$  is a hydrocarbon group of monovalence of the carbon numbers 1-20 here, and three  $R^2$  may be the same and may differ. X shows a hydroxyl group or a hydrolytic basis, and when two or more X exists, they may be the same and may differ. As for a, 1, 2, or 3b shows 0, 1, or 2, respectively. About X in bases (SiR $^1$ ) $_2$ -X $_a$ (O) $_b$ , they may be the same and may differ, m shows an integer of 0 to 19, however — what satisfies a+b=1 — carrying out — a basis expressed is added.

[0098]

It is not limited but what is necessary is just a conventionally publicly known hydrolytic basis especially as a hydrolytic basis. Specifically, a hydrogen atom, a halogen atom, an alkoxy group, an acyloxy group, a KETOKISHI mate group, an amino group, a halogen atom, an amide group, an amide group, an aminoxy group, a sulfinyloxy group, an alkenyloxy group, etc. are mentioned, for example. Among these, a hydrogen atom, an alkoxy group, an acyloxy group, a KETOKISHI mate group, an amino group, an amide group, an aminoxy group, a sulfinyloxy group, and an alkenyloxy group are preferred. Hydrolysis nature is quiet and a viewpoint of handling or a conc to especially an alkoxy group is preferred.

[0099]

A hydrolytic basis and a hydroxyl group can be combined with two silicon atom in the 1-3 ranges, and (R $^1$ amides) have 1-5 preferred ranges. When a hydrolytic basis and a hydroxyl group join together in 1-3 ranges, they may be the same and may differ.

[0100]

In particular, it is a general formula (12):



( $R^1$  and X are the same as the above among a formula.) Since a reactive silicon group expressed with an integer of 1-3 is easy to resolve, a is preferred.

[0101]

As an example of  $R^1$  in the above-mentioned general formula (11) and (12), and  $R^2$ . For example, aralkyl groups, such as aryl groups, such as cycloalkyl groups, such as alkyl groups, such as a methyl group and an ethyl group, and a cyclohexyl group, and a phenyl group, and benzyl the Tert ORGANO siloxy group  $R^1$  is indicated to be by  $SiO_2$ — which is a methyl group, a phenyl group, etc. (R) $_2$ , etc. are added. Especially in these, a methyl group is preferred.

[0102]

As more concrete illustration of a reactive silicon group, a trimethoxysilyl group, a triethoxysilyl group, a triisopropoxysilyl group, a dimethoxymethylsilyl group, a dichloromethylsilyl group, and a

dichloropropoxysilyl group are mentioned.

[0103]

Especially in this invention, an organic polymer which has the silicon containing functional group (that is, number of a term of a general formula (11) is three or more) which three or more hydrolytic bases combined on silicon in an organic polymer of the (A) ingredient can be used as an ingredient.

[0104]

A hardened material which three or more hydrolytic bases had combined this (A1) ingredient on silicon, and constructed the bridge by a silanol condensation reaction of that reactive silicon group. Good stability is shown and remarkable creep resistance and an endurance improvement effect are shown as compared with a case of reactive silicon group containing organic polymer which has two or less hydrolytic bases.

[0105]

(A1) As for the number of a term of a general formula (11) of an ingredient, it is more preferred that it is 3-5, and especially 3 is preferred. Also in it, since [ that its improvement effect of the stability of a hardenable constituent of this invention, endurance, and creep resistance is especially large and ] the Tert alkoxy silyl groups has the good availability of a raw material, it is preferred. Thing of an alkoxy group of the carbon numbers 1-20 is preferred, the thing of the carbon numbers 1-10 is more preferred, and its thing of the carbon numbers 1-4 is still more preferred here. Specifically, a trimethoxysilyl group and a triethoxysilyl group are the most preferred. Hardenable may become a hardened material when a carbon number is larger than 20.

[0106]

Generally, if weight of reactive silicon group containing organic polymer in a hardenable constituent becomes low, it is known that the endurance of a hardened material obtained will fall to low. However, if an ingredient (A1) of this invention is used as a reactive silicon group containing organic polymer, high endurance is maintainable even if it makes low weight % of reactive silicon group containing organic polymer in a hardenable constituent. Therefore, five to 28% of the weight, when it is 15 to 24 % of the weight especially preferably, since a rate of an ingredient (A1) in a hardenable constituent is compatible in low cost and high endurance, it is more preferably preferred [ rate ] ten to 26% of the weight.

[0107]

Especially in this invention, an organic polymer which has the Tert alkoxy silyl groups of the carbon numbers 2-20 can be used as a (A4) ingredient in an organic polymer of an ingredient (A1). Namely, general formula (6):



(three  $R^4$  is an organic group of monovalence of the carbon numbers 2-20 independently among a formula, respectively) — an organic polymer which has a basis expressed can be used as a (A4) ingredient.

[0108]

It is known that methanol generated in connection with a hydrolysis reaction of a methoxy silyl group has peculiar toxicity of causing an obstacle of an optic nerve. On the other hand, since a carbon number of an alkoxy group which combines the (A4) ingredient with a silicon atom is 2 to 20, toxic high methanol is not contained in alcohol generated in connection with a hydrolysis reaction of a reactive silicon group, but serves as a constituent with high safety at it.

[0109]

(A4) It is preferred that it is especially 2-4, and when it is 2, it serves as ethanol, and since alcohol generated by hydrolysis has the highest safety, it is the most preferred [ as for a carbon number of  $R^4$  of a general formula (6) of an ingredient, it is more preferred that it is 2-10, and / alcohol ].

Specifically, a triethoxy silyl group is the most preferred. When a carbon number is larger than 20, while the hardenable of a hardenable constituent may become late, an anesthetic action and stimulation of alcohol to generate may be large.

[0110]

Especially in this invention, a principal chain skeleton can use as a (A5) ingredient what is polyoxymethylene in an organic polymer of the (A4) ingredient. Namely, general formula (6):





handling 1, stability 1 of a hardenability constituent obtained 1, endurance, and creep resistance \*\* to 1, dimethoxysilane is the most preferred.

[0127] Although a method of introducing into an unsaturation binding site of an organic polymer a reactive silicon group and a reactive silicon group as a synthetic method by a radical addition reaction under a radical initiator and/or radical source-to-release existence, for example, etc. are mentioned, it is not limited in particular. As an example of a compound of having said silylalkyl group and a reactive silicon group. For example, although gamma-mercaptopropyltrimethoxysilane, gamma-mercaptopropylmethyl dimethoxysilane, gamma-mercaptopropylmethyl diethoxysilane, etc. are raised, it is not limited to these.

[0128] As mentioned above, as for a silane compound which three hydroxylic bases have combined with one silicon atom, such as trimethoxysilane, disopropoxysilane may advance. As for trialkoxysilane in a silicon capacity carbon number, such as methoxysilane, there are an alkyl group (methyl group) of 1, a dimethyl group, and a diisopropyl group. If these are combined, the above-mentioned (J) compound (like 1) which is dimethoxysilane will arise. However, such disproportionation advances in neither gamma-mercaptopropyltrimethoxysilane nor gamma-isocyanate propyltrimethoxysilane. For this reason, when using the Tori alkyl alkyl groups which has methoxy groups, such as a trimethoxysilyl group, as a silicon content group, it is preferred to use a synthetic method of (\*\*) or (\*\*).

[0130] As how to obtain an organic polymer which has the silicon content group combined with a methoxy group, it is one method of the above-mentioned (\*\*), (\*\*), and (\*\*), and a reactive silicon group is a general formula (6):



A compound (J) which has at least one methoxy group which can carry out an ester exchange reaction after obtaining an organic polymer (setting it blocked the above-mentioned (J) ingredient) which has a basis expressed with R<sup>1</sup> in formula is the same as the above). It is a general formula (7) carrying out an ester exchange reaction under existence of a transesterification catalyst or non-aqueous (10):



(Among a formula, 3-R<sup>1</sup> is an organic group of monovalence of the carbon numbers 2-20 independently, respectively, and f shows 1, 2, or 3.) — a method of manufacturing an organic polymer which has a basis expressed can be mentioned. An organic polymer which has a basis expressed with a general formula (10) shows fast curability rather than an organic polymer which has a basis expressed with a general formula (6).

[0131] In said manufacturing method, after introducing a reactive silicon group especially by a method of (\*\*), by carrying out an ester exchange reaction to the above-mentioned (J) ingredient, A method of manufacturing an organic polymer which has a basis expressed with a general formula (10). Without a disubstituted compound like dimethoxysilane by a disproportionation arising in the middle of manufacturing, there are few bad smells, and also since it becomes a good hardenability constituent of workability by hypodermally rather than an organic polymer obtained by a method of (\*\*), it is more desirable than an organic polymer obtained by a method of (\*\*).

[0132]

As a compound (J) which has at least one above-mentioned methoxy group which can carry out an ester exchange reaction, there is no limitation in particular and various kinds of compounds can be obtained.

[0133] A compound etc. which have the silicon atom combined with methyl ester of various kinds of acid, such as methanol, carboxylic acid, and sulfonic acid, and at least one hydroxylic group (J) ingredient here can be mentioned. Since a compound which has the silicon atom united with 2-4 methoxy groups on the same silicon atom as a compound which has the silicon atom combined with said at least one methoxy group has a quick ester exchange reaction speed, it is preferred. Since especially a compound that has a silicon atom united with 2-4 methoxy groups on the same silicon atom and an amino group has a quick ester exchange reaction speed, it is preferred.

[0134] When it illustrates concretely, gamma-aminopropyl trimethoxysilane, gamma-aminopropyl methyl dimethoxysilane, gamma-(2-aminoethyl) aminopropyl trimethoxysilane, gamma-(2-aminoethyl) aminopropyl methyl dimethoxysilane, Amino group content Silane, such as gamma-omega-propyltrimethoxysilane, N-phenyl-gamma-aminopropyl trimethoxysilane, and N-benzyl-gamma-aminopropyl trimethoxysilane, can be mentioned. A denatured derivative and a condensation reaction of the above-mentioned silane compound can also use the above-mentioned silane compound as a (J) ingredient.

[0135] Since an ester exchange reaction advances also under existence of a transesterification catalyst and comparatively low temperature conditions 60 ° or less, aforementioned amino group content Silane can be formed.

As for the (J) ingredient used for this invention, it is preferred to use and carry out an ester exchange reaction is 0.1-10 copies to 100 copies of reactive silicon group containing organic polymer of the (A4) ingredient. It is preferred to use it in 1-5 copies especially. The above-mentioned (J) ingredient may be used only by one kind, and may carry out two or more kind mixing use.

[0137] An organic polymer (A) which has a reactive silicon group may have straight chain shape or branching, and the number average molecular weight is 1,000-30,000 more preferably 500 to 50,000 in polystyrenes conversion in GPC. When there is a tendency whose number average molecular weight is incoincident in respect of the extension characteristic of a hardened material at less than 500 and 50000 is exceeded, since it becomes hyperviscosity, there is an incoincident tendency in respect of workability.

[0138] A reactive silicon group may be in an end or an inside of an organic polymer chain, and may be in both. Since effective network chain density of an organic polymer component contained in a hardened material formed eventually increases especially when a reactive silicon group is in a molecular terminal, it is desirable from points, like a rubber-like hardened material of high elongation becomes easy to be obtained with high intensity.

[0139] Said dimethoxysilane series polymer is a general formula intrinsically (13):



[0141]

(Among a formula, R<sup>2</sup> is a divalent organic group and is the straight chain shape or the branching alkylene group of the carbon numbers 1-14) — it being a polymer which has a repeating unit shown, and, R<sup>2</sup> in a general formula (13) has the carbon numbers 1-14, and also preferred straight chain shape or branched state alkylene group of 2-4. As the example of this repeating unit shown by a general formula (13).









[0196]

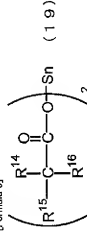
Especially in this invention, carbon of an alpha position of a carboxyl group uses carboxylic acid tin salt which is the 4th class carbon as an ingredient (C1) in carboxylic acid tin salt (Q).

SALE WHICH IS  
[0197]

(C1) As carboxylic acid tin salt of an ingredient, it is a general formula (19):

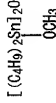
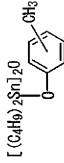
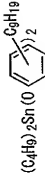
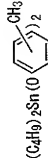
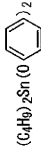
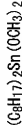
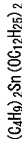
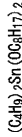
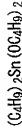
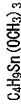
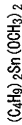
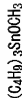
[0198]

[Formula 5]









[0225]

Although \*\* is mentioned, it is not limited to these. In these, a dialkyl tin JIARUKOKI acid is preferred. Especially the dibutyl tin JIMETOKI acid is low cost, and since it is easy to receive, it is preferred.

[0226]

(D) As amount of ingredient used, about 0.01–20 weight sections are preferred to ingredient (A1) 100 weight section. Also about 0.1–10 weight sections are preferred to ingredient (E) 100 weight section. Since the amount of ingredient used will become difficult to advance, if loadings are less than this range, it is not desirable. On the other hand, if loadings exceed this range, working life becomes short too much, and workability may worsen, and it is not desirable from a point of storage stability.

[0227]

As amount of [ in case used of using the (D) ingredient and the (C) ingredient together as a curing catalyst 1, (A1) it is preferred to consider it as (C) ingredient 0.5 – 20 weight section and (D) ingredient 0.01 – 10 weight section to ingredient 100 weight section, and also it is more preferred to consider it as (C) ingredient 1 – 10 weight section and (D) ingredient 0.02 – 5 weight section. (C) When loadings of an ingredient are less than this range, a cure rate may become slow, when loadings exceed this range, working life becomes short too much and workability may worsen. (D) The stability of a hardened material which will be obtained if an improvement effect of hardenability, depth of hardenability, an adhesive property, and thin layer hardenability may not be enough if loadings of an ingredient are less than this range, and loadings exceed this range, endurance, and creep resistance may worsen.

[0228]

The (D) ingredient can be used combining two or more sorts besides using it alone.

[0229]

In this invention, a non-tin catalyst can be used as a (E) ingredient. This non-tin catalyst has a function which improves the stability of hardened material to obtain endurance, and also improves the curing rate of hardened material with other cured hardening catalysts, when it uses as a radical condensation catalyst of an organic polymer which is an ingredient (A1) of this invention. A non-tin catalyst which is the (E) ingredient is an eco-friendly curing catalyst with high social needs.

[0230]

As a non-tin catalyst which is the (E) ingredient which can be used for this invention, although there is no restriction in particular, an organic metallic compound containing carboxylic acid, carboxylic acid metal salt other than carboxylic acid tin salt, organic sulfonic acid, alkyl acid phosphate and 3B fellows, and 4A group metal, etc. are illustrated.

[0231]

The various above-mentioned carboxylic acid which has an acid radical of carboxylic acid tin salt which is the (C) ingredient as carboxylic acid can be illustrated.

[0232]

As for said carboxylic acid, it is preferred like carboxylic acid tin salt (C) that carbon numbers including carbon of a carbonyl group are 2–20. It is more preferred that it is 6–17, and it is preferred that it is especially 8–12. A point to dicarboxylic acid or monocarboxylic acid of this ester (workability, viscosity) of dealing with it of carboxylic acid is preferred, and monocarboxylic acid is more preferred. Carboxylic acid (n-octadecanoic acid). In which said carboxylic acid is carboxylic acid (2-ethyl-2-methyl-2-octadecanoic acid etc.) and the 4th class carbon whose carbon of an alpha position of a carboxylic acid is the 4th class carbon A phthalic acid etc. are more preferred from a cure rate, but quickness of workability carboxylic acid whose carbon atom which adjoins a carbonyl group is the 4th class carbon is preferred.

[0233]

Especially as carboxylic acid, 2-ethylhexanoic acid, n-octadecanoic acid, BASA tic acid, 2,2-dimethyloctanoic acid and 2-ethyl-2,5-dimethylhexanoic acid are preferred from a point of availability, hardenability, and workability.

[0234]

As carboxylic acid metal salt other than said carboxylic acid tin salt, metal salt of the various above-mentioned carboxylic acid can be used conveniently.

[0235]



[0247]

By carboxylic acid metal salt other than carboxylic acid tin salt, organic sulfonic acid, and alkyl acid phosphate, activity is low, and when moderate hardenability is not acquired, an amine compound can be added as a co-catalyst.

[0248]

As various amine compounds, the indicated various above-mentioned amine compounds can be used as a co-catalyst of carboxylic acid tin salt (C).

[0249]

Loadings of said amine compound, about 0.01-20 weight sections are preferred to organic polymer (100 weight section of an ingredient (A1)), and also its 0.1 to 5 weight section is more preferred. A cure rate may become a little that loadings of an amine compound are less than 0.01 weight sections, and a hardening reaction becomes fully difficult to advance. On the other hand, if loadings of an amine compound exceed 20 weight sections, pot life may become short too much and is not preferred from a point of workability.

[0250]

As a metal system compound of non-tin, besides carboxylic acid metal salt other than said carboxylic acid tin salt, An organic metallic compound containing 3B fellows and 4A group metal is raised, and although a titanate organic compound, an organoaluminum compound, an organic zirconium compound, an organic boron compound, etc. are preferred from a point of activity, it is not limited to these.

[0251]

As said titanate organic compound, tetraisopropyl titanate, *tert*-butyl titanate, tetramethyl titanate, *tert*-butyl titanate, titanate compound, such as titanium chelate, such as titanium alkoxides, such as triethanolamine titanate, titanium triacetylacetonate, titanium ethylacetylacetonate, ethylene glycolate, and titanium lactate, etc. are raised.

[0252]

As said organoaluminum compound, aluminum isopropylate, aluminum alkoxides, such as mono sec-butoxy aluminum diisopropylate and aluminum sec-butoxylate. Aluminum chelate, such as aluminum triacetylacetonate, aluminum triacetylacetonate, and diisopropyl aluminum ethylacetylacetonate is

[0253]

As said zirconium compound, zirconium tetra isopropylate, zirconium alkoxides, such as a zirconium tetra-*n*-butyrate and zirconium normal butyrate, zirconium chelate, such as zirconium tetra acetylacetonate, zirconium monoacetyl acetate, zirconium bisacetylacetonate, zirconium acetylacetonate bis-ethylacetylacetonate, and zirconium acetate, is raised.

[0254]

Although \*\*\* concomitant use is also possible so, these titanate organic compounds, an organoaluminum compound, an organic zirconium compound, an organic boron compound, etc., it is desirable in a viewpoint which can reduce the amount of catalyst used especially according to concomitant use with said amine compound or an alkyl-acid-phosphate compound since it is possible to improve activity, and more desirable in a viewpoint of adjustment of working life in hardenability and ordinary temperature in an elevated temperature.

[0255]

(E) As amount of ingredient used, about 0.01-20 weight sections are preferred to ingredient (A1) 100 weight section, and also about 0.5-10 weight sections are preferred. Since a cure rate may become slow and a hardening reaction will become fairly difficult to advance if loadings are less than this range, it is not desirable. On the other hand, if loadings exceed this range, working life becomes short (too much), and workability may worsen, and it is not desirable from a point of storage stability.

[0256]

The (E) ingredient can be used combining two or more sorts besides using it alone.

[0257]

In this invention, a minute hollow body can be used as a (F) ingredient. While improving the workability (\*\*\*\*\*, thiophony) of a constituent, notably as indicated to JP H11-35922A or JP H11-31072A (if this minute hollow body is used, it is known that a weight saving of a constituent and low-costing are possible. However, it is known that the stability of a hardened material of a hardenability

constituent and endurance which are obtained will fall according to an addition of this minute hollow body.

[0258]

This hardenability constituent, which added a minute hollow body of the (F) ingredient by using an organic polymer which is an ingredient (A1) of this invention as a polymer component can maintain highly the stability of a hardened material and endurance which are obtained, improving workability (\*\*\*\*\*) notably.

[0259]

A very small hollow body (one-fourth a balloon) which is the (F) ingredient of this invention is a hollow body by which a diameter was preferably constituted from material of minerals of 500 micrometers or less or quality of organic (F) 1 mm or less as indicated, for example on "state-of-the-art of a functional filler" (CMC). Of an ingredient in particular is not limited but it is [Ingredient.] usable in various kinds of publicly known balloons.

[0260]

As for average particle density of a balloon, it is preferred that it is 0.01-1.0g/cm<sup>3</sup>; it is more preferred that it is 0.05-0.7g/cm<sup>3</sup> and it is preferred that it is especially 0.1-0.5g/cm<sup>3</sup>. If tonnage strength of a hardened material may fall if average particle density is less than this range, and average particle density exceeds this range on the other hand, a workability improvement effect may not be enough.

[0261]

An inorganic system balloon is more preferred than a point of stability and endurance to an organic system balloon.

[0262]

As said inorganic system balloon, can illustrate a silicic acid system balloon and a non-silicic acid system balloon, fly ash balloons, etc. can illustrate an alumina balloon, a zirconia balloon, a carbon balloon, etc. on a non-silicic acid system balloons. As an example of these inorganic system balloons, as a mill balloon, a win light by JICHI Chemicals, As a SANDI light by Sand Engineering Co., Ltd. and glass balloons, KARIN by Nippon Sheet Glass Co., Ltd. The Sumitomo 3M cell star Z-28, MICRO BALLOON made from EMERSONCUMING, CELAMOD GLASSMODULES made from PITTSBURGE CORNING, AS GLASS BUBBLES made from 3M, FLUIDA tube made from Fuji SHIRASHA Chemicals, and a silica balloon, as G-CEL by Asahi Glass Co., Ltd. SAURSA made from Fuji SHIRASHA Chemicals, and fly ash balloons, CEROSOL by BASF AG, and METEORING PLUTON by BASF AG. ZIRCONIUM SPIRALS made from BASF AG, BW by Shikoku KK, and KKK and KKK, and HOLLOW ZIRCONIUM SPIRALS made from ZIRCOA, KUREKASA fair made from Kurachi Chemicals and product are lots fair made from GENERAL TECHNOLOGIES are marketed as a carbon balloon.

[0263]

A balloon of thermosetting resin and a balloon of thermoplastics can be illustrated as said organic system balloon. On a thermosetting balloon, a phenol balloon, an epoxy balloon, and a urea balloon can illustrate a sarin balloon, a polystyrene balloon, a polymethacrylate balloon, a polyvinyl alcohol balloon, and a styrene acrylic balloon at a thermoplastic balloon. A balloon of thermoplastics which constructed the bridge can also be used. A balloon after foaming may be sufficient, and a balloon here is made to foam, after blending a thin containing a foaming agent, and is good also as a balloon.

[0264]

As an example of these organic system balloons, as a phenol balloon, Union Carbide UCAR and PHENOLIC MICROBALLOONS. As a urea balloon, ECOSPHERES made from EMERSONCUMING. As a polystyrene balloon, SARAN MICROSPHERES made from DOW CHEMICAL. Expanded made from Japanese filament, the Matsumoto Yushi-Seiyaku, Matsumoto microsphere. As a polystyrene balloon, DYLITE EXPANDABLE POLYSTYRENE made from ARCO POLYMERS, SBX83 by Japan Synthetic Rubber Co., Ltd. (P) are marketed by EXPANDABLE POLYSTYRENE SBX83 made from BASF WYANDOTTE. And constructed type styrene acrylic acid balloon of a bridge.

[0265]

The above-mentioned balloon may be used alone, and two or more kinds may be mixed and it may be used. The surface of these balloons Fatty acid, fatty acid ester, rosin. What was processed in order















methylsilicate 51 which were used in working example 3-4 are a condensate of a tetraethoxysilane and a tetramethoxy silane, respectively, and showed the especially outstanding effect.

(Synthetic example 2)

Using polypropylene glycol of the number average molecular weight 2,000 [about] as an initiator, and the hydroxyl group and propylene oxide of the number average molecular weight 14,500 [about] produced by polymerizing propylene oxide in the zinc hexa cyanocobaltate styra complex compound catalyst is used, AlM and polypropylene oxide was obtained in the same procedure as the synthetic example 1.

To this, AlM and polypropylene oxide, in the same procedure as the synthetic example 1, it was made to react to trimethoxysilane and the polyoxasilane series polymer (A-2) which has an average of 1.5 trimethoxysilane groups at the end was obtained.

[0324]

(Synthetic example 3)

To the allyl end polypropylene oxide obtained in the synthetic example 2, in the same procedure,

### the synthetic example

polymer (A-3) which has an average of 1.5 triethoxy silyl groups at the end was obtained.

[0325]

(Synthes

To the allyl and polypropylene oxide obtained in the synthetic example 2, in the same procedure as the synthetic example 1, it was made to react to methyl dimethoxysilane and the polyoxalkylene

series p

obtained.  
[0326]  
(The reference, examples 5-11 and comparative examples 3-5)  
Organic polymer (A-2-4) 100 weight section which has the reactive silicon group obtained in the

Organic synthesis

colloid calcium carbonate (product made from

Titanium

the amount part of thixotropic agent (made in [ Kusumoto Chemicals ], DISUPARON 6500)

duplexes

absorbent (made in Tiba Specialty Chemicals.) Tinuvin 3271 weight section, antioxidant (product







組成 (重量部)		反応性ケイ素基の構造		参考例		比較例	
有機重合体	(A4) 成分	A-10	トリメチルシリル基	100	100	9	10
		A-1	トリメチルシリル基	100	100	9	10
充填材		Winofil SPM		120	120	120	120
可塑剤		RFK-2		20	20	20	20
チクソ性付与剤		Cravallac super		50	50	50	50
光安定剤		#1-MLS-770		5	5	5	5
紫外線吸収剤		FXE Y327		1	1	1	1
酸化防止剤		15545SP		1	1	1	1
脱水剤		A-171		2	2	2	2
接着性付与剤		A-1100	トリメチルシリル基	3	3	3	3
硬化触媒		A-1120	トリメチルシリル基	2	2	2	2
還元率		材料A-U-220		87	85	25	x
クリーフ (せん断)				○	○	○	○
皮張時間				20	15	5	15
				(min)	(min)	(min)	(min)

[0955]

If the aminosiilane which has a triethoxy silyl group which is the (C) ingredient as an adhesive grant

agent is combined with the end of the (A4) ingredient using the polymer which has a triethoxy silyl group as an organic polymer as shown in the reference example 17 of Table 5. Exceeding in stability and creep resistance, change of the skinning time in storage order is small, and storage stability is good.

[0956]

The reference example 18 and the comparative example 11-12. One of the polymers (A-2) 100 weight parts which has the triethoxy silyl group obtained in the synthetic example 2 according to the combination formula shown in Table 6, as DDP20, weight section and a dehydrator — triethoxysilane (made in a oil coat) the N-beta-(aminomethyl)-gamma-aminopropyl trimethoxysilane (the Nippon Unicar make) which is the (H) ingredient as the amount part of ethyl silicate 28 duplex, and an adhesion grant agent A-1120 or N-beta-(aminomethyl)-gamma-aminopropyl triethoxysilane (the Shin-Etsu Chemical make). It added, KBE-4033 weight transformation make, neo SUTAN U-220 duplex were sealed in the glassware which carried out the nitrogen purge, and 1 liquid mold-curing nature constituent was obtained. In the comparative example 11, the leather-covered time test was performed under 50% of 23 ° humidity RH conditions, without the comparative example 12, after promoting the ester exchange reaction between reactive silicon groups by recuperating oneself for seven days at 50 ° in these 1 liquid mold-curing nature constituents, the leather-covered time test was performed under 50% of 23 ° humidity RH conditions. A result is shown in Table 6.

[0957]

[Table 6]

組成（重量部）		反応性ケイ素基の構造		参考例	比較例			
有機重合体（A4）成分	A-2	シリキシル基	100					
可塑剤	DIDP					30		
脱水剤	エチレングリコール-28					2		
接着性付与剤（H）成分	A-1120	シリキシル基	3					
硬化触媒	KBE-603		シリキシル基	2	2			
50℃×7日の養生				有	無	有		
皮張時間				(min)		3	13	12

[0058]

As shown in the reference example 18 of Table 6, the polymer which has a triethoxy silyl group is used for the end of the (A4) ingredient as an organic polymer. If the aminoalkane which has a methoxy silyl group which is the (H) ingredient as an adhesive grant agent is combined and an ester exchange reaction is promoted by cure of heat, the hardenability of an organic polymer can be raised notably.

(The reference examples 19-20 and comparative example 13)

The polyarylene ether (A-10) 100 weight section which has the reactive silicon group obtained in the synthetic example 10. Surface treatment colloid calcium carbonate (product made from Shiraiishi industry, Hakuska COR) 120 weight section, Titanium oxide (Shiraiishi Sanevo make, TIPAQUE R-820) 20 weight section, DDP55 weight section, the amount part of thixotropic grant agent (made in [Kusumoto Chemicals 1], DISUPARON 6500) duplexes, and light stabilizer (the Sankyo make), Triavin LS7701 weight section and an ultraviolet ray absorbent (made in [The Specialty Chemicals], Triavin SP1) weight section, The amount part of dehydrator vinyltrimetoxysilane (Nippon Unicar make, A-171) duplexes, adhesion grant agent N-bis-(aminoethyl)-gamma-aminopropyl trimethoxysilane (the Nippon Unicar make), A-1120) Three weight sections and the various below-mentioned curing drying conditions, it sealed in the dampproof container and 1 liquid mold-curing nature constituent was obtained. The raw decanoic acid (the product made from Japan epoxy resin) which is a non-tin catalyst of the (E) ingredient and a curing catalyst what carried out concomitant use addition of BASA tick 106 weight section and the amine (Wako Pure Chemical Industries make, lauryl amine) 0.75 weight section — the reference example 19 and isopropoxy titanium bis (diethylacetatoate) (the Matsumoto Trading make) What carried out Opa Chicks 10-150 8.5 weight section addition was made into the reference example 20. What carried out the same procedure as the reference example 19, but the polyarylene ether (Japanese east transformation make, neo SUTAN U-220) duplex was made into the comparative example 13.

As a result of measuring the recovery by the same method as the above-mentioned using these class products, the hardened material of the reference example 19 and the reference example 20 showed the recovery higher than the hardened material of the comparative example 13.

[0381]

(Synthetic example 11)  
Use polyoxypropylene glycol of the molecular weight 2,000 [about] as an initiator, and the SANOFYL group end polypropylene oxide of the number average molecular weight 25,500 [about] produced by polymerizing propylene oxide in the zinc hexa oxyacacablate glyme complex compound catalyst is used. Allyl and polypropylene oxide was obtained in the same procedure as the synthetic example 1. To this allyl and polypropylene oxide, in the same procedure as the synthetic example 1, it was made to react to triethoxysilane and the polyoxyalkylene series polymer (A-11) which has an average of 1.5 triethoxy silyl groups at the end was obtained.

[0382]

(Synthetic example 12)  
To the allyl end polypropylene oxide obtained in the synthetic example 11, in the same procedure as the synthetic example 1, it was made to react to methyl trimethoxysilane and the polyoxypropylene series polymer (A-12) which has an average of 1.5 methyl dimethoxy silyl groups at the end was obtained.

[0383]

(The reference example 21 and the comparative examples 14-15)  
Organic polymer (A-11, A-12) 95 weight section which has the reactive silicon group obtained in the synthetic example 11 and the synthetic example 12. Surface treatment colloid calcium carbonate (product made from Shiraiishi industry, Hakuska COR) 60 weight section, Surface treatment colloid calcium carbonate (product made from Shiraiishi industry, BISUKO light R) 60 weight section, Hevly calcium-carbonate (product made from Shiraiishi calcium, HOWATON SB) 20 weight section, DOP40 weight section, epoxy system plasticizer (New Japan Chemical make, SANSO size EP-S) 20 weight section, thixotropic grant agent (made in [Kusumoto Chemicals 1], DISUPARON 305) 3 weight section,

and a photo-setting resin (the Toagosei make), ARONIKUSU M-3053 weight section, light stabilizer (Sanryo make, SANORULST701) 1 weight section, Ultraviolet ray absorbent (made in [The Specialty Chemicals], thurvin 327) 1 weight section, Zero copy of minute hollow body (the product made from the Fuji SHIRISHA chemicals, the FUJIBA line H-40 which is antioxi-dant (made in [The Specialty Chemicals], IRUGA NOX 1010) 1 weight section and the (F) ingredient, or 20 copies were added, respectively, and it then kneaded with a 3 point roll, and was considered its base resin. After that added 20 copies of minute hollow bodies was made into the reference example 21, using (A-12) as an organic polymer. What added zero copy of minute hollow body was made into the comparative example 14, using (A-12) as an organic polymer, and what added 20 copies of minute hollow bodies was made into the comparative example 15, using (A-11) as an organic polymer.

[0364] Using the mixture of 2-ethylhexanoic acid in [Japanese east transformation make, U-28] (divalent) 3 weight section, amine (Wako Pure Chemical Industries make, lauryl amine) 0.75 weight section as a hardening agent, above-mentioned base resin and hardening agent were mixed uniformly, and workability (\*\*\*\*\*), and endurance were evaluated.

[0365] The constituent of the reference example 21 had workability better than the comparative example 14, and its endurance was better than the comparative example 15.

[0366] (The reference example 22 and the comparative example 16)

Organic polymer (A-1) 95 weight section which has the reactive silicon group obtained in organic polymer (A-10) 70 weight section, which has the reactive silicon group obtained in the synthetic example 10, or the synthetic example 1, Surface treatment colloid calcium carbonate (product made from Shiraishi industry, Hakukenka COR) 80 weight section, Surface treatment colloid calcium carbonate (product made from Shiraishi industry, BUSUKO light R) 60 weight section, Heavy-calcium-carbonate (product made from Shiraishi industry, HOWATON SD) 20 weight section, DOP-40 weight section, epoxy system plasticizer (New Japan Chemical make, SANSON sizer EP-S) 20 weight section, thixotropic grant agent (made in [Kusamoto Chemicals], DISUPARON 305) 3 weight section, and a photo-setting resin (the Toagosei make), ARONIKUSU M-3093 weight section, light stabilizer (Sanryo make, SANORULST701) 1 weight section, Ultraviolet ray absorbent (made in [The Specialty Chemicals], thurvin 327) 1 weight section were measured, respectively, and it then kneaded with a 3 point roll, and was considered as base resin. What added 95 copies of (A-1) as an organic polymer was made into the reference example 22, and what added 95 copies of (A-1) as an organic polymer was made into the comparative example 16. As a hardening agent, to this base resin, the mixture of 2-ethylhexanoic acid in [Japanese east transformation make, U-28] (divalent) 3 weight section, amine (Wako Pure Chemical Industries make, lauryl amine) 0.75 weight section was added, and the recovery was measured to it.

[0367] The constituent of the reference example 22 showed the recovery higher than the comparative example 16, stopping weight % of an organic polymer low.

[0368] (The reference example 23 and the comparative example 17)

Organic polymer (A-10) 95 weight section which has the reactive silicon group obtained in the synthetic example 10, Surface treatment colloid calcium carbonate (product made from Shiraishi industry, Hakukenka COR) 80 weight section, Surface treatment colloid calcium carbonate (product made from Shiraishi industry, BUSUKO light R) 60 weight section, Heavy-calcium-carbonate (product made from Shiraishi industry, HOWATON SD) 20 weight section, DOP-40 weight section, thixotropic system plasticizer (New Japan Chemical make, SANSON sizer EP-S) 20 weight section, thixotropic grant agent (made in [Kusamoto Chemicals], DISUPARON 305) 3 weight section, and a photo-setting resin (the Toagosei make), ARONIKUSU M-3093 weight section, light stabilizer (Sanryo make, SANORULST701) 1 weight section, Ultraviolet ray absorbent (made in [The Specialty Chemicals], thurvin 327) 1 weight section, Antioxidant (made in [The Specialty Chemicals], IRUGA NOX 1010) 1 weight section and zero copy of epoxy resin (the product made from Japan epoxy resin, Epicoat 828), or five copies were measured, respectively, and it then kneaded with a 3 point roll, and

was considered as base resin. What added five copies of epoxy resins was made into the reference example 23. What added zero copy of epoxy resin was made into the comparative example 17. As a hardening agent, to this base resin, the mixture of 2-ethylhexanoic acid in [Japanese east transformation make, U-28] (divalent) 3 weight section and amine (Wako Pure Chemical Industries make, lauryl amine) 0.75 weight section was added, and the recovery was measured to it.

[0369] The constituent of the reference example 23 showed the recovery higher than the comparative example 17.

[0370] (The reference example 24 and the comparative example 18)

Organic polymer (A-10) 95 weight section which has the reactive silicon group obtained in the synthetic example 10, Surface treatment colloid calcium carbonate (product made from Shiraishi industry, Hakukenka COR) 80 weight section, Surface treatment colloid calcium carbonate (product made from Shiraishi industry, BUSUKO light R) 60 weight section, Heavy-calcium-carbonate (product made from Shiraishi industry, HOWATON SD) 20 weight section, DOP-40 weight section, epoxy system plasticizer (New Japan Chemical make, SANSON sizer EP-S) 20 weight section, thixotropic grant agent (made in [Kusamoto Chemicals], DISUPARON 305) 3 weight section, and a photo-setting resin (the Toagosei make), ARONIKUSU M-3093 weight section, light stabilizer (Sanryo make, SANORULST701) 1 weight section, Ultraviolet ray absorbent (made in [The Specialty Chemicals], thurvin 327) 1 weight section and antioxidant (made in [The Specialty Chemicals], IRUGA NOX 1010) 1 weight section were measured, respectively, and it then kneaded with a 3 point roll, and was considered as base resin.

[0371] 2-ethylhexanoic acid in [Japanese east transformation make, U-28] (divalent) 3 weight section and amine (Wako Pure Chemical Industries make, lauryl amine) 0.75 weight section and thixotropic grant agent (made in [Kusamoto Chemicals], DISUPARON 305) 3 weight section, and the mixture of 2-ethylhexanoic acid in [Japanese east transformation make, U-28] (divalent) 3 weight section as a hardening agent is made into the reference example 24, the thing using the mixture of 2-ethylhexanoic acid in [Japanese east transformation make, U-28] (divalent) 3 weight section and amine (Wako Pure Chemical Industries make, lauryl amine) 0.75 weight section as a hardening agent was made into the comparative example 18. Base resin and a hardening agent were mixed uniformly and the recovery and the layer hardenability were evaluated.

[0372] The constituent of the reference example 24 showed good thin layer hardenability rather than the comparative example 18, while the high recovery was shown.

[0373] (Synthetic example 13)

To the allyl end and polyisobutylene obtained according to the example of manufacture of JP-H1-200639A, under existence of Pt catalyst, it was made to react to triethoxysilane and the polyisobutylene (A-13) which has a triethoxy allyl group at the end was obtained.

[0374] (Synthetic example 14)

To the allyl end and polyisobutylene obtained in the synthetic example 13, under existence of Pt catalyst, it was made to react to methyl dimethoxysilane and the polyisobutylene (A-14) which has a methyl dimethoxy allyl group at the end was obtained.

[0375] (Reference example 25 and the comparative example 19)

To organic polymer (A-13, A-14) 100 weight section which has the reactive silicon group obtained in the synthetic example 13 and the synthetic example 14, the amount part of divinylbenzene bisacrylate (Japanese east transformation make, neo SUTAN U-220) duplex was added, and the hardened material was obtained. The thing using (A-13) as an organic polymer was made into the reference example 25, and the thing using (A-14) was made into the comparative example 18. The hardened material of the reference example 25 showed the recovery higher than the comparative example 19.

[0376] (Synthetic example 15)



